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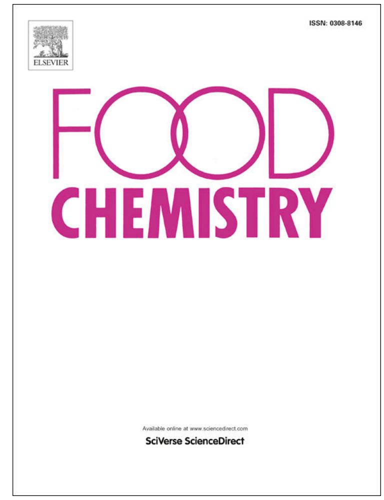

Aroma generation in sponge cakes: The influence of sucrose particle size and sucrose source

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Abstract

The influence of sucrose source and particle size was investigated in relation to the volatile and aromatic properties of sponge cakes. Six sponge cake formulations were studied using two sucrose sources (sugarbeet and sugarcane), at two particle sizes (large and small) with controls. Volatiles profiles and odour active compounds were identified by gas chromatography mass spectrometry and olfactometry. Sixty two volatile compounds were identified, incorporating twenty five odour active compounds /co-eluting compounds, with 5 odours perceived without any corresponding volatile. Particle size had the greatest impact on volatile abundance, with particle size especially influencing pyrazine abundance. Five odour active volatiles (methional, furfural, 2,3-dimethylpyrazine, heptanal and (E)-2-octenal) contributed most to the aroma of these sponge cakes. Small particle size particularly from sugarbeet yielded higher levels of some Maillard and caramelisation reaction compounds, such as furfural (spicy / bready), where larger particle size supressed volatile abundance in comparison to the control.

Keywords: Bakery, volatile compounds, reformulation, odour active, gas-chromatography-olfactory

1. Introduction

The growing demand for food products formulated with minimal ingredients, and the expectations of food manufactures to refrain from the use of artificial additives and preservatives, challenges the conventional approach to new product development, especially in relation to sugar and fat reduction. Sucrose reduction is a primary objective for food manufacturers globally in an attempt to tackle the burden of health-related chronic disease, such as obesity and type II diabetes. Polyols and artificial sweeteners were once considered solutions to sucrose reduction in sugar laden food and beverage matrices (Ghosh & Sudha, 2012; Struck et al., 2014) however, a substantial shift towards clean-label formulation has occurred due to consumer demand (Asioli et al. 2017).

Baked confectionery products are of interest reformulation, especially in relation to sucrose reduction (Garvey et al., 2020a; Luo et al., 2019; Sahin et al., 2019). Clean-label sucrose reduction has previously been employed in sponge cakes utilising apple pomace powder, whey permeate powder and oligofructose (Garvey et al., 2020b; Milner et al., 2020). Sponge cakes formulated with apple pomace powder and oligofructose resulted in increased 'off-flavour' (apple pomace powder) and more pronounced 'toasty', and 'nutty' attributes, than the control. A recent study by Maruyama et al. (2021), highlighted that consumers identified sugarcane as the most 'natural' ingredient across a range of different raw materials, with fructose classed as the least 'natural' in the sugar category.

Sucrose, otherwise known as sugar (or table sugar), is traditionally derived from sugarbeet (*Beta vulgaris*) and sugarcane (*Saccharum officinarum*), is available as a refined, granulated sugar with a sucrose content of ≥ 99%. Although the sugars are considered as identical in terms of chemical composition, differences in their sensory characteristics have been documented. Urbanus, Cox, Eklund, Ickes, Schmidt, and Lee (2014) using descriptive analysis noted that sugarbeet was characterised by 'off-dairy', 'oxidized', 'earthy', and 'barnyard' aromas, whereas sugarcane was profiled as 'fruity' in mouth aroma, with a sweet aftertaste. Similarly, Urbanus, Schmidt, and Lee (2014) found the inclusion of sugarbeet with sugarcane created an 'off-aroma' in sugar syrup and pavlova, where the volatile fraction of raw sugarbeet juice was shown to contain 2-methoxy-3-(1-methylpropyl)pyrazine, dimethyl disulfide, and geosmin (Pihlsgård et al., 2000), with geosmin postulated to be the biggest contributor to 'off' aromas (Freidig & Goldman, 2014). Freidig & Goldman (2014) characterised sugarcane as containing primarily alcohols (1-pentanol, 2-butanol, 2-octanol) that may potentially influence flavour depending upon their abundance and odour activity. Differences in the thermal behaviour of sugarbeet and sugarcane have also been identified (Lu et al., 2017), which may have an impact on how these sugars behave when used in baked confectionery products from a sensory point of view, due to the formation of odour active volatiles from Maillard reactions (MR) and caramelisation reactions (CR).

Richardson et al. (2018) investigated manipulating sugar crystal size in chocolate brownies, experimenting with small (459-972 μm), medium (627-1214 μm) and large particle (924-1877 μm) fractions. The authors identified that smaller sugar crystal particles yielded a chocolate brownie with statistically higher 'sweet taste' compared to that of the control (200-5181 μm) in a consumer study and postulated that small sugar particles could be employed to reduce overall sugar content in baked confectionery products, whilst retaining the fundamental sensory appeal. Although manipulation of ingredient particle size (PS) has resulted in baked confectionery products with similar sensory properties as traditional formulas, there is potential for the aroma profile to be altered. The PS of sucrose and cocoa liquor mixture for chocolate manufacture, has been shown to influence the release of 'characteristic chocolate' aroma compounds; 3-methylbutanal, 2-phenylethanol, furfuryl alcohol, acetic acid, methylpyrazine, 2,3-dimethylpyrazine, 2,5-dimethylpyrazine and trimethylpyrazine, when analysed by gas chromatography-olfactory (GC-O) (Afoakwa et al., 2009).

Most volatiles identified by gas chromatography mass spectrometry (GC-MS) cannot influence sensory perception, as they are not present at concentrations above their odour thresholds. GC-O is a technique often used in association with GC-MS to identify the impact of individual volatile compounds influencing sensory perception using trained (sniffers) panellists and aroma extraction dilution techniques. GC-O was successfully employed to investigate the influence of sucrose replacers on the odour active compounds of clean-label sponge cakes (Garvey et al., 2020c). To the best of our knowledge, no study has been previously conducted to explore the influence of sugar particle size on the volatile aroma compounds of sponge cakes. The objective of this study was to use a similar approach to Garvey et al. (2020c) to investigate the influence of sucrose source (SS); (sugarcane and sugarbeet), and PS on the volatile aroma profile of sponge cakes. Volatile analysis was conducted using an optimised headspace solid phase micro-extraction gas-chromatography mass-spectrometry (HS-SPME-GS-MS) method (Garvey et al., 2020c), and by GC-O.

2. Materials and methods

2.1 Analytical standards

Olfactory training standards were of analytical grade; ethyl butyrate, octanal, p-cresol, and dimethyl disulphide and heptanal of ≥99% and ≥95% purity respectively (Merck Ireland, Arklow, Co. Wicklow, Ireland), were prepared at 0.3% (w/v) in methanol and stored at −18 °C until required. For each GC-O training session, a stock solution was diluted to 0.03% (w/v) in distilled water to allow the odours to be of adequate potency, as described by Garvey et al. (2020b).

2.2 Sucrose samples

Sugarbeet sugar samples (refined sucrose 99.7%, PS = 0.45 – 0.65 mm) were purchased in a local supermarket in the form of granulated sugar, under the brand name Siucra (Nordzucker, Germany). Sugarcane sugar (raw sucrose, sucrose content not declared, PS= 0.3 – 0.9 mm) was sourced from Equal Exchange (Sunderland, UK). Sugar was stored in airtight plastic containers at ambient temperatures of 20 °C prior to sieving. For effective sieving, sugar was dried at 70 °C for 1 h in an oven (Binder, ED 115, Germany) to reduce moisture content.

2.3 Sugar sieving

The dried sugar was sieved using a laboratory test sieve shaker (Octagon 200 test sieve shaker, Endecotts Ltd, London, UK) using a range of sieve sizes (125, 250, 500, 710 and 1800 µm). To increase the amount of small PS retained in a given sieve, and to reduce the labour of manually grinding the sugar crystal particles, a portion of the sugars were blitzed in a blender (NutriBullet 600, Australia) to expedite the generation of small PS. All sugars were sieved in batches of 200 g at an 8-mm amplitude for 5 min. The resultant sieved sugars were separated into, PS ≤ 250 µm, referred to as small PS (SPS), and ≥ 710 µm, referred to as large PS (LPS). Each fraction was stored in airtight plastic containers until required for baking.

2.4 Particle size distribution

The PS distributions (PSD) of each powder fraction were analysed in triplicate, measured by laser light scattering using a Mastersizer 3000 (Malvern Instruments Ltd., UK), equipped with an Aero S dry powder dispersion unit. PS measurements were expressed as smallest PS (D10), mean PS (D50) and largest PS (D100), corresponding to the maximum diameters of 10%, 50%, and 90% of the particles, respectively (in % of total volume).

2.5 Sponge cake preparation

The sponge cake formulation and baking conditions was as described by Garvey et al. 2020c, utilising the Control recipe, with the fractionated sugar substituted equally for sucrose quantity, as seen by Richardson et al. (2018). The fractionated sugar crystal particles were assigned to six sponge cake formulations (Table 1): sugarbeet control (SB-C), sugarcane control (SC-C), sugarbeet SPS (SB-SPS), sugarcane SPS (SC-SPS), sugarbeet LPS (SB-LPS) and sugarcane LPS (SC-LPS). All sponge cake formulations consisted of identical formulas and independent baking trials were carried out in duplicate.

Plain flour (200 g) (Odlums, Ireland) and baking powder (4 g) (Dr. Oekter, UK) were sifted into a bowl followed by the addition of the trial sugar (Súcra, Nordzucker, Germany; Equal Exchange, UK), cake margarine (90 g) (Stork, UK), free range eggs (90 g) (local retailer), and water (70 g). The contents were mixed using a household mixer (Kenwood Mixer, Model KMM710, UK) at speed 1 for 30 seconds, scraped and mixed again for a further 2 min at speed 2. Miniature loaf tins (80 mm × 60 mm × 40 mm) were filled with 80 g of cake batter and baked at 180 °C for 45 min in a domestic convection oven (Zanussi, Bedfordshire, UK). The cakes were left to cool and placed in sealed storage bags until subsequent HS-SPME-GS-MS or GC-O analysis (which took place within 24 hours after baking).

2.6 Volatile analysis

Volatile analysis was carried out as described by Garvey et al. (2020c). Each sponge cake was sliced vertically and 1cm of the outer crust was removed. Cake crumb (3 g) was added to an amber 20 ml screw capped headspace vial (Apex Scientific Ltd, Co. Kildare, Ireland) and equilibrated for 5 min, at 60°C with pulsed agitation for 5 seconds at 350 rpm, using the Gerstal MultiPurpose Sampler (GMPS) agitator/heater. Volatile analysis was carried out utilising a GMPS rail system (Anatune, Cambridge CB3 0NA, UK) connected to a Shimadzu GP2010 plus gas chromatograph (GC) (Mason Technology Ltd, Dublin, Ireland) using headspace solid-phase microextraction (HS-SPME). The SPME fibre; 30/50 µm DVB/CAR/PDMS (Agilent Technologies Ltd., Ireland), was exposed to the headspace above the samples, at a depth of 21 mm, for 60 min at 60 °C. The fibre was retracted, injected into the GC inlet and desorbed for 3 min at 250 °C, followed by 3 min at 270 °C in GMPS fibre bake-out station, to minimise the carryover of compounds. Each sponge cake formula was analysed in triplicate.

2.7 Identification of odour active compounds by gas chromatography-olfactometry

HS-SPME-GC-O analysis was undertaken on all experimental samples. Sponge cake formulations were sliced and blitzed in a food processor (NutriBullet 600, Australia) to combine cake crust and crumb uniformly to represent a masticated sample. Ten (sniffer) assessors were chosen for the GC-O analysis based on their performance in an olfactory assessment using three different Sniffin' Stick tests (identification, discrimination, and threshold) (Hummel et al., 1997). Prior to sample analysis, the panellists were exposed to a standard stock solution (as described in 2.1), designed for GC-O training, comprised of 5 compounds; dimethyl disulphide ('sulphur','decomposing'), ethyl butyrate ('fruity', 'pineapple'), heptanal ('fatty', 'green'), octanal ('orange', 'fruity') and ρ-cresol ('barnyard'). This step allowed panellists to familiarise themselves with the GC-O process and software, as well as the range of odours they could potentially encounter during the GC-O analysis of sponge cake formulations. Formulations were evaluated in a randomized incomplete block design, allowing for each sample to be evaluated 5 times. If an aroma was detected by at least 3 out of 5 evaluations, it was established as odour active (Feng et al. 2015).

Volatile extraction was carried out by the HS-SPME procedure (as previously described). GC-O analyses was performed on an Agilent 7890 GC with a flame ionization detector, 5973 N mass detector (Agilent Technologies, Ltd, Cork, Ireland), and an Gerstel ODP-3 olfactory detector port (Anatune Ltd, Cambridge, UK). The volatile compounds were separated on DB-624 UI (20m x 1.8mm x 1 μm) (Agilent Technologies Ltd, Ireland) column. Eluting compounds were split 1:1:1 into the MS detector, flame ionisation detector and the sniffing port simultaneously by means of a column flow splitter. The carrier gas was helium, held at a pressure of 9.8 psi and a flow rate of 1.209 mL /min. GC conditions consisted of an initial oven temperature of 80 °C, held for 2 min and increased at 10 °C/min to 220°C. The GC run time was shortened to 21 min to reduce the risk of assessors experiencing fatigue during a sniffing session (however it still encompassed the volatile range of interest). In addition, the transfer line to the sniffing port was conditioned with humidified air to reduce olfactory fatigue and prevent the occurrence of condensation droplets collecting in the nasal port. The ion source temperature was 220 °C and the interface temperature was set at 260 °C. The MS mode was electronic ionization (70 eV) with the mass range scanned between m/z 35-250. Compounds were identified using mass spectra comparisons to the NIST 2014 mass spectral library, comparison of LRI to the mid polar column from the previous analysis and to standards where possible. Spectral de-convolution was also performed to confirm identification of compounds using Automated Mass Spectral Deconvolution and Identification System (AMDIS).

To determine the threshold at which each odour active compound could be perceived, Aroma Extraction Dilution Analysis (AEDA) was carried out by manipulation of the GC injection split ratio (Cepeda-Vázquez et al., 2018). Formulations were analysed at a splitless (1:1 ratio), and for subsequent analysis, the operating mode of GC analysis was changed from splitless to split injection and the split ratio was adjusted to 1:10, 1:20, 1:50, 1:100, 1:150, 1:200, and 1:250, allowing for adequate dilution to determine the most odour active compounds in the formulations. For this task, three sniffer assessors were chosen from the five previously selected for the AEDA portion of the study, with each split ratio analysed in duplicate. The last split ratio at which a compound could be detected was referred to as the factor dilution (FD) for that compound.

2.8 Statistical Analysis

Data analysis was handled accordingly based on the normality of the data. To identify differences in area values of the volatile compound data, a Welch test was applied, with difference in means identified by Games Howell post hoc test, working at an alpha level of 0.05. A two-way analysis of variance (ANOVA) was applied to volatile compound data to identify the main effects of independent variables sucrose source and sugar PS on the area peak value of volatile compounds (dependent variable). A two-way ANOVA was also employed to identify any interaction effect amongst the independent variables. Statistical analysis was performed using IBM SPSS Statistics 24 for windows (SPSS Inc., IBM Corporation, NY, USA). Principle component analysis (PCA) was constructed using the “factoextra” and “Facto- MinoR” packages in R (v 3.4.1, R Foundation for Statistical Computing, Vienna, Austria).

3. Results and Discussion

3.1 Particle size distribution

PS diameter means of the control sugars were 601 µm (SB-C) and 657 µm (SC-C) and demonstrate a similar distribution range of 52-1630 µm (SB-C) and 46- 1850 µm (SC-C) (Figure 1). The SPS particles, which were subjected to sieving and grinding also had a similar distribution ranges of 3.5-586 µm (SB-SPS) and 1.1-666 µm (SC-SPS), with the following mean diameters 198 µm (SB-SPS) and 178 µm (SC-SPS). The largest disparity was identified in the LPS sucrose samples obtained using the 710 µm sieve. The distribution range for SC-LPS was 45.6 - 3000 µm, with 29.9% of the SC-LPS particles at < 500 µm, whereas, SB-LPS ranged from 352 – 2100 µm. Diameter means for SB-LPS and SC-LPS were 867 µm and 1100 µm, respectively.

3.2 Volatile profile of sugarcane and sugarbeet sponge cakes

3.2.1 Influence of sugar source on volatile formation

Volatile analysis identified a total of 62 volatile compounds across all formulations (Table 2), with ketones, aldehydes, furans, pyrazines, as the main chemical classes. Principle component analysis (PCA) was conducted to identify any associations between individual volatiles and the different sponge cake formulations (Figure 2). Figure 2 depicts the PCA plot of each sponge cake formulation and the volatiles which accounted for 77% of the total variance amongst the formulations. It is easy to see that most volatiles are more associated with the controls (SB-C and SC-C) and the SB-SPS sponge cake samples. Based on results of a two-way ANOVA (Table 2), overall, as an independent variable SS did not confer a significant effect on volatile formation, with the exception for toluene ($P = 0.001$). Toluene was significantly ($P < 0.05$) higher in SB-C, compared to SC-C. This compound was highest in SB-SPS, however, it is unlikely to influence sensory perception due to an extremely low odour activity.

3.2.2 Influence of sucrose particle size on volatile formation

Sugar PS had a significant influence on 20 of the 62 volatile compounds in these sponge cakes, but especially pyrazines as all of those detected were impacted by PS. The abundances of pyrazines were greater in SB-C, SC-C, SB-SPS and SC-SPS than SB-LPS and SC-LPS ($P < 0.001$), except for 2-ethyl-3,6-dimethylpyrazine (Table 2). Control and SPS size formulas were particularly high in 2,5-dimethylpyrazine, trimethylpyrazine and 2-ethyl-5-methylpyrazine, which may also be linked to Strecker degradation, compared to LPS sponge cakes (Table 2). Afoakwa et al. (2009) also found that the inclusion of large particle sizes (sucrose and cocoa liquor mix) in chocolate formulation decreased some pyrazines, such as methylpyrazine, 2,3-dimethylpyrazine, 2,5-dimethylpyrazine, trimethylpyrazine. α-Dicarbonyl compounds are also produced through sugar fragmentation / dehydration during MR and CR and play a role in pyrazine formation (Deeth & Lewis, 2017). It is possible that the decreased surface area, due to the LPS, decreased the generation of α-dicarbonyl compounds, leading to less pyrazines in the LPS sponge cakes.

The abundance of heptanal and (E)-2-nonenal were significantly lower ($P < 0.001$) in SB-LPS and SC-LPS than the controls (SB-C and SC-C), with (E)-2-octenal (although this was also lower in SC-SPS than in SC-C) and decanal also lower ($P < 0.001$) in SB-LPS compared to SB-C (Table 2). Even though (E,Z)-decadienal was significantly different ($P < 0.001$) due to PS, only SC-SPS was significantly lower ($P < 0.001$) than the SC-C. Thus, it would appear that some lipid oxidation reactions may be retarded by the inclusion of LPS particles, due to lower abundances of many aldehydes generated by lipid oxidation in comparison to the controls. The generation of furans was significantly increased in Robusta coffee when PS were reduced to 200 μm – 300 μm, compared to larger fractions of coffee > 425 μm (De Vivo et al., 2022). In this study, only furfural and acetylfuran abundances were also significantly lower ($P < 0.001$) in SB-LPS and SC-LPS than in SB-C and SC-C (Table 2). SB-LPS had significantly lower levels of 2-furanmethanol, compared to SB-SPS. However, abundances of ethyl furan were significantly lower in SB-SPS in comparison to SB-LPS. Although sucrose will not accelerate furan formation as strongly as reducing sugars, such as glucose, fructose and lactose etc. in baked goods (Wang et al., 2009; Zhang et al. 2012), the inclusion of SPS induces a lower melting point (Richardson et al., 2018), which may accelerate the hydrolysis of sucrose, resulting in more MR and CR, and thus yield higher levels of some furans. The presence of higher levels of 2-acetylfuran in SB-SPS and SC-SPS sponge cakes may also indicate greater hydrolysis as it is also derived from glucose (Kocadağlı et al., 2020). Both carbon disulfide and 2-acetlthiazole were also impacted by PS (P < 0.001), however abundance levels of carbon disulfide varied considerably within replicates and only SB-LPS were significantly lower than SB-SBS. For 2-acetylthiazole only SB-LPS was lower than the SB-C, and SC-SPS lower than SC-C.

Overall the inclusion of LPS (SB-LPS and SC-LPS) had the least influence on aroma volatile formation in comparison to the controls (SB-C and SC-C) than SPS (SB-SPS and SC-SPS), with the exception of some methyl ketones, and ethanol (Figure 2, Table 2). The methyl ketones, 2-heptanone, 2-octanone, and 2-nonanone were present at higher levels in LPS formulas, particularly in SB-LPS. On heating, methyl ketones can be formed by the decarboxylation of β-keto fatty acids, or the β-oxidation and decarboxylation of free saturated fatty acids (Pasqualone et al., 2014). Therefore, the LPS appears to positively influence these reactions. As stated, ethanol was present in greater amounts in the LPS formulas and may be as a result of heightened microbiological activity (Martin & Ames, 2001). However, the high volatility of ethanol makes it difficult to analyse by HS techniques, a factor to consider in relation to the interpretation of these results.

3.2.3 Influence of the interactive effect of sucrose source and particle size on volatile formation

A significant (P < 0.001) interaction effect was also identified, between SS and PS, for a number of compounds; acetone, 2-butanone, 2-methylpropanal, 3-methylbutanal, heptanal, (E)-2-octenal, (E)-2-nonenal and ethyl furan (Table 2). The aldehydes; 2-methylpropanal and 3-methylbutanal, were significantly (P < 0.001) influenced by the interaction of SS and PS, with SB-SPS and SC-C having significantly (P < 0.05) higher levels of 2-methylpropanal and 3-methylbutanal compared to the other formulations (except SC-LPS, P > 0.05). 2-Methylpropanal and 3-methylbutanal derive from Strecker degradation reactions of amino acids, valine and leucine, respectively, and are identified as important outputs of MR, with sugar form (mono-, di- saccharide) an influential factor in their generation (Palmonari et al., 2020). It is difficult to discern how SS and

PS interacted in the formation of these compounds, as no distinct trend is evident (Table 2). Levels of the aldehydes heptanal, (E)-2-octenal ($P < 0.001$), and (E)-2-nonenal (P 0.001) were also influenced by the interaction between SS and PS. Acetone (2-propanone) and 2-butanone were also influenced by the interaction of SS and PS ($P < 0.001$), although acetone was only statistically lower in SC-SPS in comparison to SC-C, and 2-butanone was not statistically different in relation to PS. Acetone and 2-butanone are products of carbohydrate metabolism likely produced by the yeast, but may also be impacted by the baking process. 2-Butanone, was identified as having a 'butterscotch', 'artificial', 'vanilla' aroma in liquid sugarbeet (Pihlsgård, et al., 2000) and has also been suggested to arise from MR and CR reactions in baked confectionery products (Pasqualone et al., 2015; Valli et al., 2012). Ethyl furan was also influenced by the interaction of SS and PS ($P < 0.001$), and as previously stated was also influenced by PS. It is difficult to discern the true influence of the interactive effect of SS and PS on the generation of these compounds.

3.3 Influence of sugar particle size and source on the odour active volatile profile of sponge cakes

Thirty distinct odour active compounds (Table 3) were identified in all sponge cakes by olfactometry, the identity of 25 confirmed through comparison of molecular ion matching and RI index's. Co-elution of aroma compounds is common, and in this study, benzaldehyde co-eluted with 1-octen-3-ol, 2-ethyl-5-methylpyrazine with trimethylpyrazine, phenylacetaldehyde with 3-ethyl-2,5-dimethylpyrazine, and furaneol with 2-nonanone, as previously identified by Garvey et al. (2020c). The aroma descriptions of 6 unknown compounds were also included (Table 3). The unknown compounds were odour active enough to be perceived, but below the limits of detection by the mass spectrometer, which is also not uncommon in GC-O analysis.

It is apparent that both SS and PS influenced the odour profile of these sponge cakes (Figure 3). The odour activities of twelve of the thirty volatiles were perceived similarly for both SB-C and SC-C formulations (Table 3), which demonstrates that the impact of PS was greater than SS, in agreement with the volatile analysis. The aromatic profiles of the sponge cakes consisted of the following compounds; methional, furfural, heptanal, 2,3-dimethylpyrazine, 2,5-dimethylpyrazine, (E)-2-octenal, 2-nonanone/ furaneol, 2-acetylpyrrole and 2,3-dihydro-3,5-dihydroxy-6-methy-4(H)-pyran-4-one, with these compounds identified in at least one formula at an FD of 50, or higher.

Comparing the odour activity of SB to SC sponge cakes, SB-C sample had greater odour intensity for (E)-2-octenal, 2,3-dimethylpyrazine, furaneol / 2-nonanone phenylacetaldehyde, 2,3-dihydro-3,5-dihydro-6-methyl 4(H)-pyran-4-one, 2-acetylpyrrole, 1-octen-3-ol / benzaldehyde, acetophenone and 2-ethyl-5-methyl-pyrazine / trimethylpyrazine (Table 3). While 2,5-dimethylpyrazine, 2-methylpropanal, pentylfuran and 3,5-octadien-2-one were perceived at greater intensity in the SC-C samples, compared to SC-B.

Comparing the influence of PS in sugarbeet formulas, it is apparent that modifying PS altered the odour profile, as the FD values of all compounds except 2,3-butanedione and 3-methylbutanal were altered. The potency of methional and (E)-2-octenal were reduced on the inclusion of SPS and LPS, reducing the FD from 200 to 150, for both SB-SPS and SC-LPS. Similar trends were noted for changes in PS, for example, the following volatiles had similar FD values in SB-C and SB-SPS but decreased in SB-LPS; heptanal 2,5-dimethylpyrazine, 2,3-dihydro-3,5-diydroxy-6-methyl-4(H)-pyran-4-one, 2-furanmethanol, phenylacetaldehyde, nonanal. As the majority of these compounds are products of MR, it demonstrates a reduction of some MR compounds by the inclusion of LPS particles, and therefore a reduction of the impact of some characteristic sponge-cake aroma compounds. Neither 2-methylpropanal, or acetol, were perceived in SB-C or SB-SPS but were detected in SB-LPS.

Similarly, the odour intensity of 2,3-dimethylpyrazine was influenced by PS, with the FD values compared to the control increasing in SB-LPS, yet decreasing in SB-SPS. This result does not reflect the values identified in Table 2, however, this is most probably due to the sporadic nature of pyrazine formation in the crust, from accelerated MR and CR, hence leading to a higher prevalence of pyrazines. Paraskevopoulou et al., (2012) identified 2,3-dimethylpyrazine as a key odour active compound in the crust of wheat bread, and wheat bread formulated with lupin protein isolate, compared to the crumb.

Furfural is a major contributor to the aroma of sponge cakes (Garvey et al. 2020c), with the odour activity significantly influenced by SPS, evident by an increase in the intensity of furfural in SB-SPS compared to SB-C and SB-LPS, reflecting the levels identified in Table. 2. Other furanic compounds, 2-acetylfuran and pentylfuran, were also influenced by PS manipulation, with both acetylfuran and pentylfuran not perceived in SB-SPS (matching the volatile data where lower levels of these furanic compounds were noted). The odour activity of furaneol / 2-nonanone and 2-acetylthiazole displayed higher FD values in SB-C and SB-LPS, compared to SB-SPS. LPS particles had a prominent influence on the odour activity of 2-acetylpyrrole, exhibiting a 'cotton candy' aroma up to an FD of 100. Acetylpyrrole is proposed to be generated through intact sugar fragments during MR (Totlani & Peterson, 2007), indicating its irregular generation in cake crust, as also identified by Garvey et al. (2020c).

The odour profile of the SC formulations appears the most impacted by PS. SC-C and SC-LPS were dominated largely by 'potato-like' methional (Figure 3), however a significant decrease was identified in SC-SPS with a drop to FD 50. Lower values for methional were also identified in the volatile fraction of the crumb of SC-SPS, although not significant, it may relate to the varying levels in the odour active profile. Methional is derived from Strecker degradation of the amino acid methionine and has also previously been identified with a similar 'potato' odour in sponge cakes (Boeswetter et al., 2019; Garvey et al. 2020c), and is considered an important contributor to wheat bread crust (Pu et al., 2019; Whitfield & Mottram, 1992).

Similar to the trend in the SB formula, furfural contributed significantly to the profile of all SC sponge cakes. Furfural appears to play an important role in the desired aroma of sponge cakes and its prominent aroma is due to the favoured generation pathway of 1,2-enolization that takes place during baking (Zhang et al., 2012). Heptanal 'fatty, oily' contributed to the odour of SC-C, however, it was only perceived at an FD of 10 in SC-SPS and SC-LPS. This occurrence is difficult to interpret, due to the significantly higher levels of heptanal identified in the volatile fraction of the crumb (Table 2). However, heptanal is a product of lipid oxidation deriving from the auto-oxidation of oleic acid (Zamora & Hidalgo, 2005) and it appears that PS not only influences MR and CR (Matsakidou et al., 2010) but also lipid oxidation. Garvey et al. (2020c) previously identified heptanal as strongly influencing the aroma of sponge cakes formulated with caster sugar.

(E)-2-Octenal 'earthy, damp' also significantly contributed to the aroma of all formulations except for SC-SPS and has also been previously shown to influence the aroma of sponge cakes formulated with clean-label ingredients (Garvey et al. 2020b). The potency of this compound across sponge cake formulas is likely related to its origin; generated from LO reactions of unsaturated fatty acids (Whitfield & Mottram, 1992). However, (E)-2-octenal seems to be suppressed in SPS formulas, where it is evident that more MR and CR are occurring. SC-SPS contained high levels of 'nutty, bready' 2,5-dimethylpyrazine (FD 100), which may be explained by heightened MR in the crust, although, this was not reflected in the crumb. Similar behaviour was seen by 2,3-dihydro-3,5-dihydro-6-methyl 4(H)-pyran-4-one. These Strecker derived compounds have been identified as key contributors to the crust and crumb of sponge cakes (Pozo-Bayón et al., 2007).

4. Conclusions

SS and PS and their interactive effect had a significant interaction effect on the abundance of several volatile compounds. The impact PS was much more significant than with either SS or the interactive effect of SS and PS, and was especially evident in relation to pyrazines. When SPS were incorporated into the sponge cake formulas, particularly from SB, both MR and CR were accelerated based on the enhanced formation of thermally derived volatiles. This is presumably due to the lower melting point of SPS particles. The impact of LPS was significantly less than SPS in relation to the volatile profile, although for both types of sucrose (SB and SC) levels of methyl ketones and ethanol increased, indicating that their generation pathways are directly affected by sucrose PS. GC-O identified thirty odour active compounds in these sponge cake formulations, with methional, furfural, heptanal, 2,3-dimethylpyrazine and (E)-2-octenal having the greatest contribution to the overall aroma. The FD value of compounds differed as a result of formulation changes in SS and PS, with the volatile profile of the cake crust differing between cake formulas, and therefore influencing aroma profiles. To the best of our knowledge, no study has been conducted exploring the influence of sugar PS on the volatile aroma compounds of sponge cakes. This study suggests small sucrose particles have potential to produce higher levels of 'bready / spicy' furans, 'cake crust' pyrazines and 'fatty / oily' heptanal in sponge cakes. This approach is likely to be applicable to other baked products using granulated sugar, however the specific impact is likely to be impacted by individual formulations and baking parameters. Further work is required to understand the difference in sensory perception of these formulas to fully validate sucrose PS manipulation as a tool to aid in reformulation of reduced sucrose sponge cakes.

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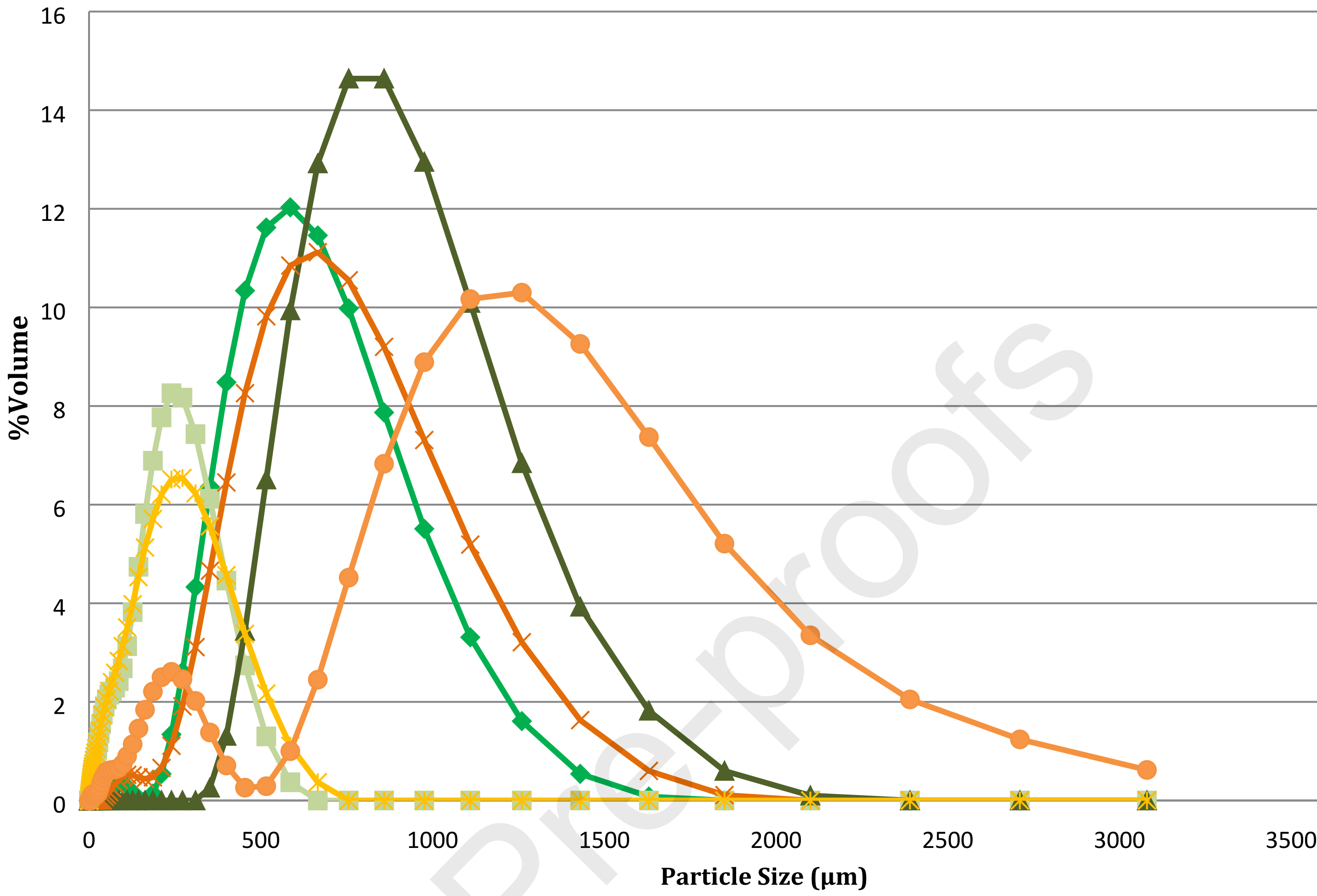
TITLE: Aroma generation in sponge cakes: The influence of sucrose particle size and sucrose source
Food Chemistry Submission- Figures

Figure 1. Particle size distribution of experimental sugars.

Figure 2. Principle component analysis of the volatile compounds associated with sugarbeet control (SB-C), sugarcane control (SC-C), sugarbeet small particle size (SB-SPS), sugarcane small particle size (SC-SPS0, sugarbeet large particle size (SB-LPS) and sugarcane large particle size (SC-LPS) sponge cakes analysed by headspace solid-phase microextration gas chromatography mass spectrometry.

Figure 3. Odour active profiles of Sugarbeet; SB-C (A), SB-SPS (B) and SB-LPS (C), and Sugarcane; SC-C (D), SC-SPS (E) and SC-LPS (F) sponge cakes. Pie chart segments represent the dilution factor values of the main odour active compounds with larger segments indicating compounds perceived at higher dilutions (Table 4), with compounds from ≥FD 50 only. Colour chart reflects the volatile compounds present in the pie charts



16
14
12
10
8
6
4
2
0
%Volume
0
500
1000
1500
2000
2500
3000
3500
Particle Size (µm)

Figure 1

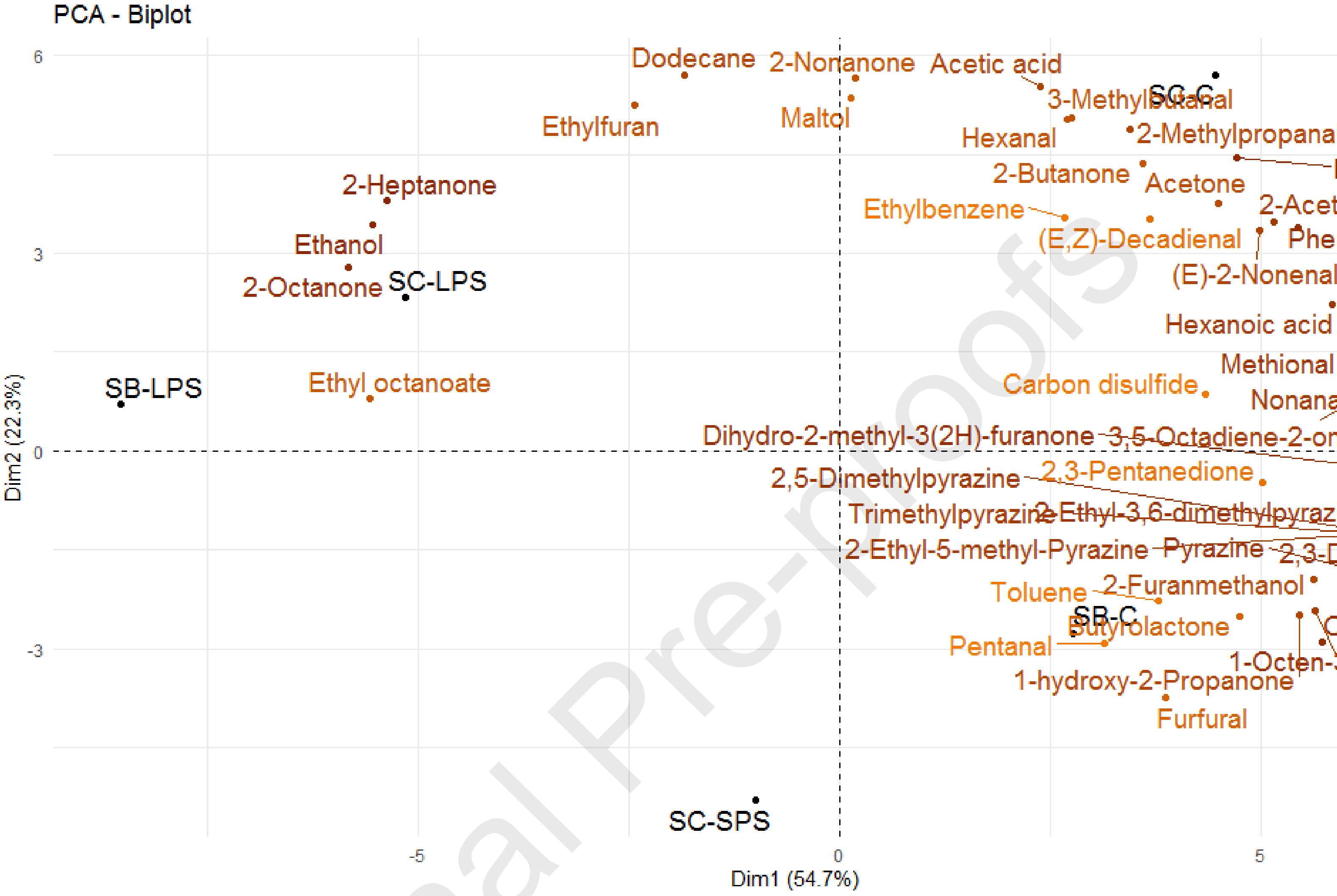

PCA - Biplot
Dim1 (54.7%)
Dim2 (22.3%)
SB-LPS
SC-LPS
SC-SPS
SC-C
SB-C
Dodecane
2-Nonanone
Acetic acid
Ethylfuran
Maltol
3-Methylbutanal
Hexanal
2-Methylpropanal
2-Heptanone
2-Butanone
Acetone
Ethylbenzene
Ethanol
(E,Z)-Decadienal
2-Octanone
(E)-2-Nonenal
Hexanoic acid
Methional
Ethyl octanoate
Carbon disulfide
Dihydro-2-methyl-3(2H)-furanone
2,5-Dimethylpyrazine
2,3-Pentanedione
Trimethylpyrazine
2-Ethyl-5-methyl-Pyrazine
Pyrazine
Toluene
2-Furanmethanol
Butyrolactone
Pentanal
1-hydroxy-2-Propanone
Furfural

Figure 2

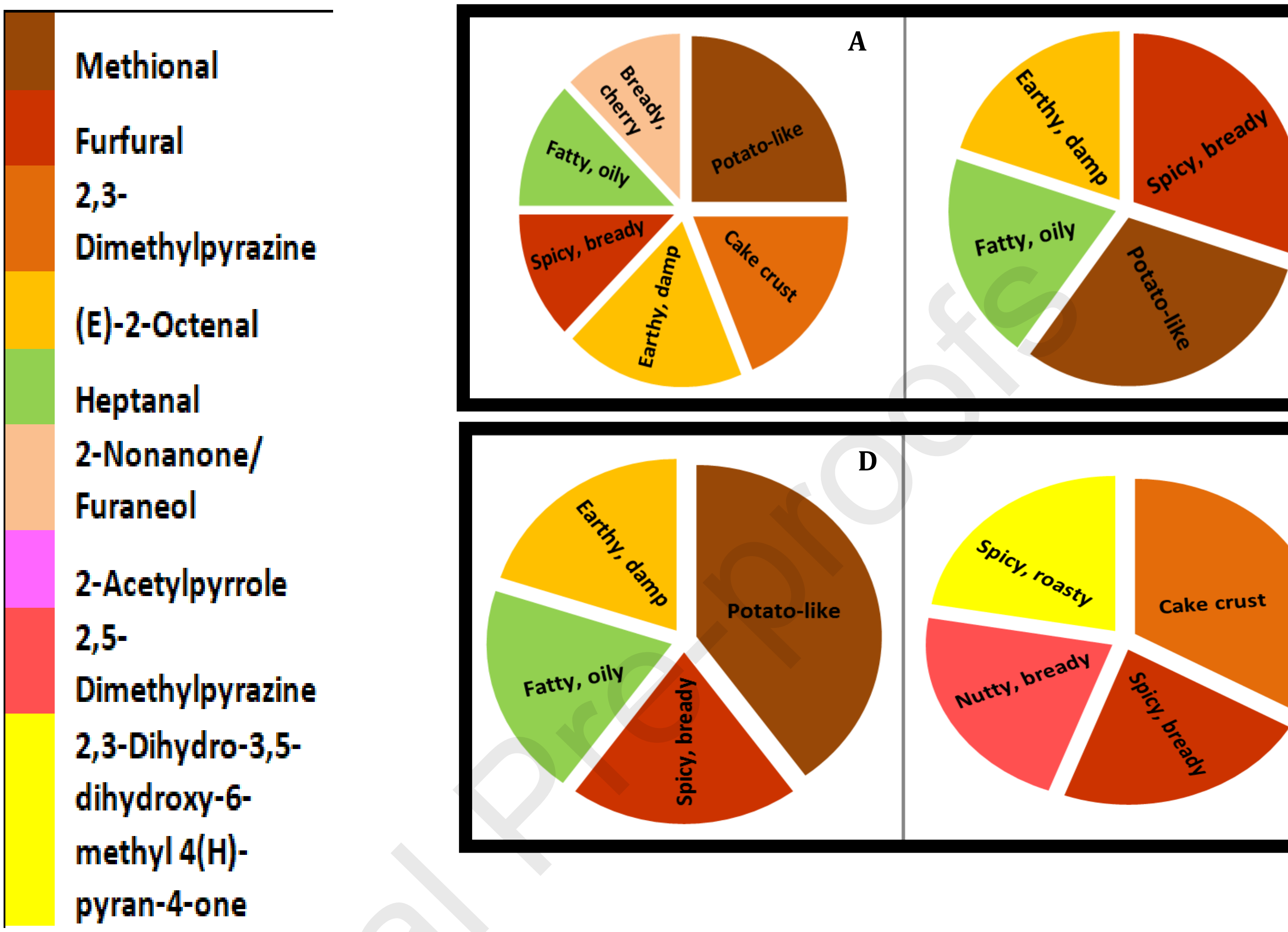


Figure 3

TITLE: Aroma generation in sponge cakes: The influence of sucrose particle size and sucrose source
Food Chemistry Submission- Tables

Table 1. Summary of experimental sugars, particle sizes and respective sponge cakes

Sucrose Source (SS)	Sucrose Particle Size (SPS)	Respective Sponge Cake	Abbreviation
	< 250 µm	Sugar Beet- Small Particle Size	SB-SPS

Beet	52-1630 µm	Sugar Beet- Control	SB-C
	>710 µm	Sugar Beet- Large Particle Size	SB-LPS
Cane	< 250 µm	Sugar Cane- Small Particle Size	SC-SPS
	46- 1850 µm	Sugar Cane- Control	SC-C
	>710 µm	Sugar Cane- Large Particle Size	SC-LPS

Table 2. Average (n=6) peak area values of volatile compounds identified in sugarbeet control (SB-C), sugarcane control (SC-C), sugarbeet small particle size (SB-SPS), sugarcane small particle size (SC-SPS), sugarbeet large particle size (SB-LPS) and sugarcane large particle size (SC-LPS) sponge cakes analysed by headspace solid-phase microextration gas chromatography mass spectrometry.

npound	**Sugarbeet**			**Sugarcane**			**Two-way ANOVA (α=0.001)**	
	SB-C	**SB-SPS**	**SB-LPS**	**SC-C**	**SC-SPS**	**SC-LPS**	**Interaction Effect (SS x SPS)**	**Sucros source**
	0.44 ± 0.2 ^{ab}	0.92 ± 0.3 ^{ab}	0.39 ± 0.1 ^{b}	0.85 ± 0.1^{a}	0.42 ±0.1 ^{b}	0.65 ± 0.3 ^{ab}	**<0.001**	**n.s**
e	8.34 ± 1.6 ^{ab}	9.49 ± 1.0 ^{a}	8.56 ± 0.3 ^{ab}	8.81 ± 1.0 ^{ab}	7.67 ± 0.6 ^{b}	9.67 ± 1.7 ^{ab}	**n.s**	**n.s**
	0.18 ± 0.04 ^{b}	0.50 ± 0.3 ^{ab}	0.24 ± 0.021 ^{ab}	0.49 ± 0.2 ^{a}	0.22 ± 0.02 ^{ab}	0.35 ± 0.1 ^{ab}	**<0.001**	**n.s**
	0.20 ± 0.1	0.24 ± 0.1	0.30 ± 0.1	0.21 ± 0.1	0.13 ±0.1	0.16 ± 0.1	**n.s**	**n.s**
ne	8.58 ±8.6 ^{ab}	7.3 ± 0.7 ^{a}	5.80 ± 0.4 ^{b}	8.43 ± 1.47 ^{a}	7.64 ± 4.94 ^{ab}	6.65 ± 1.54 ^{ab}	**n.s**	**n.s**
ropanone	34.06 ± 17.7	65.63 ± 31.2	12.18 ± 3.8	26.94 ± 14.1	37.60 ± 28.4	14.41 ± 11.2	**n.s**	**n.s**
	37.36 ± 9.2 ^{bc}	34.18 ± 5.1^{c}	59.03 ± 4.4 ^{a}	48.67 ± 3.6 ^{ab}	39.24 ± 11.1^{bc}	58.93 ± 13.3^{ab}	**n.s**	**n.s**
ne	5.91 ± 1.3	6.29 ± 1.5	1.27 ± 1.9	2.80 ± 2.6	3.72 ± 2.8	3.06 ± 2.3	**n.s**	**n.s**
	42.94 ± 10.5^{ab}	38.74 ± 5.0 ^{b}	58.32 ± 1.6 ^{a}	45.81 ± 3.8 ^{b}	41.56 ± 11.4 ^{ab}	53.45 ± 11.9 ^{ab}	**n.s**	**n.s**
	34.71 ± 7.0 ^{ab}	34.69 ± 4.3 ^{b}	42.16 ± 1.1 ^{a}	54.71 ± 25.6 ^{ab}	33.94 ± 3.2 ^{b}	38.42 ± 5.0 ^{ab}	**n.s**	**n.s**
	0.57 ± 0.1 ^{a}	0.91 ± 0.4 ^{a}	0.18 ± 0.01 ^{b}	0.59 ± 0.2 ^{a}	0.34 ± 0.1 ^{ab}	0.26 ± 0.2 ^{ab}	**n.s**	**n.s**
2-one	0.97 ± 0.2 ^{ab}	1.21 ± 0.3 ^{a}	0.61 ± 0.01 ^{b}	1.28 ± 0.8 ^{abc}	0.91 ± 0.4 ^{abc}	0.55 ± 0.04 ^{c}	**n.s**	**n.s**
e	7.22 ± 1.4	7.55 ± 1.1	7.29 ± 0.4	7.34 ± 1.2	6.24 ± 0.4	6.77 ± 0.4	**n.s**	**n.s**
nal	0.39 ± 0.1 ^{b}	0.75 ± 0.2 ^{a}	0.38 ± 0.21 ^{b}	0.77 ± 0.2 ^{a}	0.32 ± 0.02 ^{b}	0.64 ± 0.1 ^{ab}	**<0.001**	**n.s**
al	3.43 ± 1 ^{ab}	6.34 ± 1.3 ^{a}	4.08 ± 0.3 ^{a}	6.29 ± 0.5 ^{ab}	3.13 ± 0.6 ^{b}	5.56 ± 1.7 ^{ab}	**<0.001**	**n.s**
al	4.26 ± 1.0	5.43 ± 1.0 ^{a}	3.92 ± 0.2 ^{a}	4.31 ± 1.9 ^{b}	3.00 ± 0.3 ^{b}	5.35 ± 1.7 ^{a}	**n.s**	**n.s**
	1.56 ± 0.2	1.53 ± 0.3	0.61 ± 0.5	1.32 ± 0.2	0.85 ± 0.5	0.96 ± 0.5	**n.s**	**n.s**
	22.08 ± 4.6	20.93 ± 4.2	19.45 ± 1.4	23.88 ± 2.5	17.76 ± 5.9	21.94 ± 4.8	**n.s**	**n.s**
	19.52 ± 2.4^{b}	24.74 ± 5.9^{ab}	12.84 ± 0.5^{c}	25.88 ± 1.9^{a}	14.77 ± 7.0^{abc}	14.36 ± 3.8^{bc}	**<0.001**	**n.s**
	0.38 ± 0.1 ^{a}	0.42 ± 0.2 ^{ab}	0.19 ± 0.05 ^{b}	0.39 ± 0.1 ^{ab}	0.24 ± 0.1 ^{ab}	0.28 ± 0.1 ^{ab}	**n.s**	**n.s**
	5.32 ± 1.5 ^{a}	6.72 ± 3.6 ^{ab}	2.11 ± 0.04^{b}	4.52 ± 0.7 ^{a}	3.38 ± 0.9 ^{ab}	2.91 ± 0.8 ^{b}	**n.s**	**n.s**
	3.95 ± 1.2 ^{ab}	3.93 ± 1.0 ^{a}	1.86 ± 0.1 ^{b}	3.28 ± 0.6 ^{a}	3.14 ± 1.9 ^{ab}	2.04 ± 0.2 ^{b}	**n.s**	**n.s**
	4.46 ± 0.3 ^{a}	4.74 ± 1.0 ^{a}	1.33 ± 0.1 ^{b}	4.96 ± 0.7 ^{a}	2.08 ± 1.1 ^{b}	2.82 ± 1.8 ^{ab}	**<0.001**	**n.s**
hyde	4.46 ± 0.4 ^{a}	4.68 ± 1.0 ^{ab}	3.11 ± 0.9 ^{ab}	5.48 ± 1.2 ^{a}	3.35 ± 0.6 ^{b}	3.74 ± 1.0 ^{ab}	**n.s**	**n.s**
	6.14 ± 1.7 ^{ab}	6.14 ± 09 ^{a}	4.10 ± 0.6 ^{ab}	6.64 ± 1.9 ^{ab}	4.86 ± 1.0 ^{ab}	4.14 ± 0.4 ^{b}	**n.s**	**n.s**
	0.96 ± 0.1 ^{ab}	0.90 ± 0.1^{b}	0.55 ± 0.1 ^{c}	1.31 ± 0.2 ^{a}	0.57 ± 0.2 ^{c}	0.60 ± 0.3 ^{bc}	**0.001**	**n.s**
	1.52 ± 0.6^{ab}	1.44 ± 0.3^{a}	0.51 ± 0.1^{c}	1.52 ± 0.6^{abc}	0.84 ± 0.3^{b}	0.70 ± 0.2 ^{b}	**n.s**	**n.s**
nal	2.67 ± 0.7 ^{a}	1.86 ± 0.6 ^{ab}	1.56 ± 0.7 ^{ab}	3.45 ± 1.3 ^{ab}	1.48 ± 0.3 ^{b}	1.64 ± 0.7 ^{ab}	**n.s**	**n.s**
	0.21 ± 0.1 ^{b}	0.37 ± 0.1 ^{ab}	0.44 ± 0.02^{a}	0.42 ± 0.1 ^{a}	0.25 ± 0.1 ^{b}	0.46 ± 0.03 ^{a}	**<0.001**	**n.s**
hyl-	1.76 ± 0.4 ^{a}	2.87 ± 1.2 ^{ab}	0.71 ± 0.1^{c}	2.22 ± 0.9 ^{ab}	1.72 ± 0.3 ^{ab}	0.90 ± 0.4 ^{bc}	**n.s**	**n.s**

e								
	34.59 ± 5.5 ^{ac}	46.66 ± 20.3 ^{ab}	19.74 ± 4.5 ^{b}	37.65 ± 9.4 ^{a}	60.43 ± 23.7 ^{a}	19.81 ± 8.5 ^{bc}	**n.s**	**n.s**
	0.51 ± 0.1 ^{ab}	0.61 ± 0.1 ^{a}	0.46 ± 0.03 ^{b}	0.67 ± 0.1 ^{a}	0.47 ± 0.2 ^{ab}	0.54 ± 0.1 ^{ab}	**n.s**	**n.s**
ol	18.69 ± 6.9 ^{ab}	34.98 ± 15.2 ^{a}	7.69 ± 3.0 ^{b}	24.15 ± 10.2 ^{ab}	28.45 ± 22.0 ^{ab}	8.82 ± 7.1 ^{b}	**n.s**	**n.s**
	1.79 ± 0.3 ^{a}	2.47 ± 0.8 ^{ab}	1.01 ± 0.2 ^{bc}	1.94 ± 0.5 ^{a}	2.23 ± 0.8 ^{abc}	0.99 ± 0.3 ^{c}	**n.s**	**n.s**
	84.08 ± 16.8 ^{a}	110.27 ± 13.7 ^{a}	40.22 ± 1.3 ^{b}	93.72 ± 23.7 ^{ac}	73.19 ± 45.4 ^{abc}	59.63 ± 23.3 ^{bc}	**n.s**	**n.s**
	4.18 ± 1.0 ^{a}	6.07 ± 1.9 ^{a}	2.11 ± 0.3 ^{b}	4.13 ± 0.5 ^{a}	4.32 ± 0.5 ^{a}	2.63 ± 0.8 ^{b}	**n.s**	**n.s**
e	35.16 ± 8.1 ^{a}	50.01 ± 17.6 ^{a}	16.37 ± 2.4 ^{b}	33.36 ± 4.4 ^{a}	32.79 ± 6.5 ^{a}	18.14 ± 7.0 ^{b}	**n.s**	**n.s**
yrazine	52.05 ± 12.6 ^{a}	81.82 ± 31.1 ^{a}	23.81 ± 5.2 ^{b}	50.87 ± 8.0 ^{a}	45.94 ± 14.0 ^{ab}	26.65 ± 12.0 ^{b}	**n.s**	**n.s**
e	6.71 ± 2.1 ^{a}	9.26 ± 3.1 ^{a}	2.77 ± 0.5 ^{b}	6.11 ± 1.2 ^{a}	6.26 ± 1.5 ^{a}	3.08 ± 1.3 ^{b}	**n.s**	**n.s**
yrazine	3.13 ± 1.0 ^{a}	4.64 ± 2.0 ^{ab}	1.35 ± 0.3 ^{b}	2.96 ± 0.5 ^{a}	2.76 ± 0.9 ^{ab}	1.49 ± 0.7 ^{b}	**n.s**	**n.s**
	7.28 ± 2.3 ^{a}	12.77 ± 5.6 ^{a}	2.70 ± 0.8 ^{b}	6.99 ± 1.7 ^{a}	6.39 ± 2.7 ^{ab}	3.05 ± 1.7 ^{b}	**n.s**	**n.s**
e								
zine	9.68 ± 2.7 ^{a}	16.28 ± 7.3 ^{ab}	3.78 ± 1.1 ^{b}	9.07 ± 2.2 ^{a}	8.10 ± 3.4 ^{ab}	4.15 ± 2.4 ^{b}	**n.s**	**n.s**
	0.63 ± 0.2 ^{a}	1.00 ± 0.5 ^{ab}	0.18 ± 0.2 ^{ab}	0.58 ± 0.2 ^{ab}	0.49 ± 0.2 ^{ab}	0.25 ± 0.2 ^{b}	**n.s**	**n.s**
ne								
	38.88 ± 42.6 ^{b}	41.37 ± 46.9 ^{b}	316.84 ± 34.5 ^{a}	163.56 ± 179.0 ^{ab}	86.07 ± 7.4 ^{b}	296.64 ± 10.8 ^{a}	**n.s**	**n.s**
	8.26 ± 2.5	5.47 ± 0.4	5.57 ± 1.0	4.75 ± 0.5	5.85 ± 3.7	6.34 ± 1.5	**n.s**	**n.s**
	15.06 ± 2.7 ^{a}	14.57 ± 2.2 ^{a}	7.56 ± 0.9 ^{b}	11.86 ± 2.8 ^{ab}	12.91 ± 8.6 ^{ab}	9.35 ± 2.0 ^{b}	**n.s**	**n.s**
l	0.70 ± 1.0	1.44 ± 1.3	1.01 ± 0.04	1.91 ± 0.8	1.57 ± 1.1	1.10 ± 0.1	**n.s**	**n.s**
	2.88 ± 3.1	2.73 ± 2.1	2.75 ± 0.6	4.47 ± 2.0	1.70 ± 0.8	2.57 ± 1.9	**n.s**	**n.s**
	2.55 ± 0.7 ^{a}	2.85 ± 0.9 ^{a}	0.98 ± 1.1 ^{ab}	3.05 ± 2.2 ^{ab}	0.96 ± 1.1 ^{ab}	1.07 ± 0.4 ^{b}	**n.s**	**n.s**
	52.76 ± 7.4 ^{ab}	54.04 ± 8.6 ^{ab}	60.08 ± 2.2 ^{a}	54.93 ± 9.0 ^{ab}	46.82 ± 4.3 ^{b}	51.86 ± 2.5 ^{b}	**n.s**	**n.s**
	0.56 ± 0.1	0.69 ± 0.3	0.34 ± 0.2	0.61 ± 0.2	0.75 ± 0.3	0.50 ± 0.1	**n.s**	**n.s**
	0.68 ± 0.7 ^{ab}	0.67 ± 0.7 ^{ab}	0.20 ± 0.005 ^{ab}	0.17 ± 0.2 ^{ab}	0.09 ± 0.03 ^{ab}	0.25 ± 0.1 ^{ab}	**n.s**	**n.s**
e	0.05 ± 0.1 ^{b}	0.11 ± 0.1 ^{ab}	0.22 ± 0.05 ^{a}	0.10 ± 0.1 ^{ab}	0.15 ± 0.1 ^{ab}	0.19 ± 0.1 ^{ab}	**n.s**	**n.s**
le	111.22 ± 69.3 ^{ab}	59.17 ± 23.4 ^{a}	13.17 ± 1.9 ^{b}	96.14 ± 19.4 ^{a}	50.30 ± 16.6 ^{a}	61.75 ± 34.3 ^{ab}	**n.s**	**n.s**
anhydride	0.57 ± 0.1 ^{ab}	0.59 ± 0.1 ^{ab}	0.62 ± 0.02 ^{a}	0.59 ± 0.1 ^{ab}	0.50 ± 0.05 ^{b}	0.54 ± 0.03 ^{b}	**n.s**	**n.s**
e	0.09 ± 0.1 ^{b}	0.13 ± 0.1 ^{ab}	0.16 ± 0.1 ^{ab}	0.35 ± 0.1 ^{a}	0.21 ± 0.03 ^{ab}	0.17 ± 0.1 ^{ab}	**n.s**	**n.s**
e	0.46 ± 0.2	0.72 ± 0.6	0.22 ± 0.2	0.73 ± 0.5	0.46 ± 0.4	0.26 ± 0.3	**n.s**	**n.s**
	1.46 ± 0.6 ^{ab}	1.27 ± 0.2 ^{a}	0.64 ± 0.1 ^{b}	0.68 ± 0.3 ^{b}	0.70 ± 0.1 ^{b}	0.83 ± 0.2 ^{ab}	**n.s**	**0.001**
	5.38 ± 2.1 ^{ab}	4.83 ± 0.7 ^{a}	2.47 ± 0.1 ^{b}	4.50 ± 2.6 ^{ab}	5.70 ± 3.0 ^{ab}	2.56 ± 0.8 ^{b}	**n.s**	**n.s**
le	1.49 ± 0.3 ^{ac}	1.58 ± 0.7 ^{ab}	0.79 ± 0.05 ^{b}	2.04 ± 0.2 ^{a}	1.10 ± 0.2 ^{bc}	1.36 ± 0.3 ^{ab}	**n.s**	**n.s**
	1.25 ± 0.4	1.19 ± 0.1	1.31 ± 0.05	1.41 ± 0.1	1.21 ± 0.1	1.35 ± 0.1	**n.s**	**n.s**
	1.45 ± 1.0	1.75 ± 1.3	1.90 ± 0.7	1.94 ± 1.2	1.27 ± 1.4	1.57 ± 0.9	**n.s**	**n.s**

Table 3. Odour active compounds with corresponding odour impression and factor dilution value identified in sugarbeet control (SB-C), sugarcane control (SC-C), sugarbeet small particle size (SB-SPS), sugarcane small particle size (SC-SPS), sugarbeet large particle size (SB-LPS) and sugarcane large particle size (SC-LPS). The retention times in mins of the unknown compounds is listed in brackets after each.

Volatile compound	Descriptor	Sugarbeet			Sugarcane		
		SB-C	SB-SPS	SB-LPS	SC-C	SC-SPS	SC-LPS
2-Methylpropanal	*Spicy, cake*	n.d	n.d	10	20	n.d	20
2,3-Butanedione	*Butterscotch, caramel*	20	20	20	20	1	20
3-Methylbutanal	*Toffee,sweet, butterscotch*	20	20	20	20	20	20
Acetol	*Fruity, sweet*	n.d	n.d	20	n.d	n.d	20
Unknown 1 [4.3]	*Toasty, floral*	n.d	n.d	10	n.d	n.d	20
Furfural	*Spicy, bready*	100	150	100	100	100	50
2-Furanmethanol	*Bisuit, cake crust*	50	50	n.d.	50	n.d	n.d
Heptanal	*Oily, fatty*	100	100	50	100	10	10
2,5-Dimethylpyrazine	*Nutty, bready*	20	20	10	50	100	1
2,3-Dimetylpyrazine	*Cake crust*	150	50	200	50	150	10
Methional	*Potato-like*	200	150	150	200	50	100
2-Acetylfuran	*Bready, sweet*	50	n.d	10	50	n.d	n.d
Pentylfuran *	*Bready*	1	n.d	10	10	n.d	n.d
1-octen-3-ol /Benzaldehyde	*Mushroom / sweet*	50	10/50	10/50	20	20	10/20
2-ethyl-5-methyl-pyrazine/ Trimethylpyrazine	*Sweet, cake crust, mouldy, woody, soil*	20	1	10	1	10	20
2-Acetylthiazole	*Sweet, bready, cake crust*	20	10	20	20	1	20
Phenylacetaldehyde	*Honey, floral*	50	50	10	20	n.d	10
E-2-octenal	*Earthy, damp*	150	100	100	100	50	100
Furaneol 2-Nonanone	*Cake crust, bready, sweet, cherry*	100	50	100	50	50	50
Acetophenone	*Cake crust. Toasty*	50	n.d	20	10	20	50
Nonanal	*Sweet, cake crust*	20	20	10	20	10	20
2-Acetylpyrrole	*Sweet, cotton candy*	50	1	100	20	20	20
(3,5) Octadien2one	*Caramel, cake crust, sweet*	n.d	20	10	1	10	20

Unknown 2 [9.7]	*Cake crust, bready, butterscotch*	50	1	10	1	20	20
Dodecane	*Damp, musty*	50	20	20	50	20	50
Unknown 3 [10]	*Fresh, woody, citrus*	1	1	50	50	1	1
Unknown 4 [10.40]	*Biscuit, bready, floral*	1	n.d	n.d	n.d	n.d	n.d
2,3-Dihydro-3,5-dihydroxy-6-methyl 4(H)-pyran-4-one	*Cake crust, spicy, roasty*	50	50	20	20	100	20
Unknown 5 [10.60]	*Nutty, roasty*	n.d	1	50	1	n.d	n.d
Unknown 6 [11.3]	*Tea, citrus*	n.d	n.d	1	20	n.d	n.d

Highlights

- The influence of sucrose particle size, and sucrose source on the volatile and aroma of sponge cakes was investigated.
- Sponge cakes were formulated from cane or beet sucrose at three sucrose particle sizes (small, control and large).
- Particle size had the greatest impact on overall volatile abundance and aroma properties.
- Small particle size formulas yielded higher Maillard reaction compounds (pyrazines).

Individual Contributions

Kieran Kilcawley, Maurice O'Sullivan: Conceptualization
Emer Garvey; Kieran Kilcawley, Maurice O'Sullivan: Methodology
Emer Garvey, Kieran Kilcawley: Validation
Emer Garvey; Kieran Kilcawley, Maurice O'Sullivan: Formal analysis
Emer Garvey: Investigation
Kieran Kilcawley, Maurice O'Sullivan, Joseph Kerry: Resources
Kieran Kilcawley: Data curation
Emer Garvey: Writing – original draft
Kieran Kilcawley, Maurice O'Sullivan, Joseph Kerry: Writing – review and editing
Emer Garvey: Visualization
Kieran Kilcawley, Maurice O'Sullivan, Joseph Kerry: Supervision
Kieran Kilcawley, Maurice O'Sullivan: Project administration

Maurice O'Sullivan, Joseph Kerry: Funding acquisition